IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: Takeaki SAIKI et al.

Confirmation No.: 9218

Serial No.:

10/533,169

Group Art Unit:

4171

Filed:

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Examiner:

DOLLINGER, Michael M.

For:

METHOD FOR THE PREPARATION OF A SILICON-CONTAINING

POLYSULFIDE-TYPE POLYMER

DECLARATION UNDER 37 CFR § 1.132

MAILSTOP: AMENDMENT Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450

Dear Sir:

- I, Makoto Iwai, hereby state that:
- 1. I am a citizen of Japan.
- 2. I have a Doctor of Engineering degree from Chiba University in Chiba, Japan. I am currently employed in a Senior Chemist role for Dow Corning Toray Co., Ltd. of Tokyo, Japan. I have worked in the silicone field for 25 years and I have been employed by Dow Corning Toray Co., Ltd. for the past 25 years.
- 3. I am a joint inventor of the pending U.S. Patent Application, Serial No. 10/533,169. I am a person highly skilled in the art of silicones including silicone compositions

H&H # 071051.00008 Serial No. 10/533.169 and methods for producing such silicone compositions including, in particular, siliconcontaining polysulfide-type polymers.

- 4. In the present application, the invention, which is a method for the preparation of a silicon-containing polysulfide-type polymer, includes simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond; (B) a polysulfide polymer with at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur.
- 5. I am aware of, have read, and understand the disclosure of U.S. Patent No. 3,476,826 to Millen (the '826 patent), which is entitled "ORGANO-SILANE MODIFIED POLYSULFIDE POLYMERS AS ADHESIVE ADDITIVES OR PRIMERS FOR HIGH RANK POLYSULFIDE BASED ADHESIVE COMPOSITIONS."
- 6. I am aware of, have read, and understand the disclosure of U.S. Patent No. 3,925,331 to Ely et al. (the '331 patent), which is entitled "POLYMERIC SEALANTS."
- 7. I am aware of, have read, and understand the disclosure of U.S. Patent No. 4,096,131 to Price et al. (the '131 patent), which is entitled "ONE PACK POLYSULPHIDE SEALANTS."
- 8. For the reasons described in Paragraphs 9-25 immediately below, the invention in the present application is unique and distinguishable from the '826, '331 and '131 patents, as well as combinations thereof. Specifically, upon analyzing the teachings of the '826, 331 and '131 patents, even when combined, the silicon-containing polysulfide-type

H&H # 071051.00008 Serial No. 10/533,169 polymer or the method of preparing the silicon-containing polysulfide-type polymer of the invention would not be made or otherwise envisioned.

- 9. After closely analyzing the '826 patent, I can find nothing in the '826 patent that teaches a method for the preparation of a silicon-containing polysulfide-type polymer by simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond; (B) a polysulfide polymer with at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur, as claimed for the invention.
- 10. I recognize that Example 1 of the '826 patent describes a method of preparing a vinyl triethoxy silane polysulfide by reacting a polysulfide polymer and vinyl triethoxy silane in the presence of 2,2'-azobisisobutyronitrile.
- 11. I also recognize that Example 2 of the '826 patent describes a method of preparing a high sulfur rank liquid polysulfide polymer by reacting a liquid polysulfide polymer and paraformaldehyde in the presence of triethylamine to form a blocked polysulfide polymer, and subsequently reacting the blocked polysulfide polymer with elemental sulfur, n-dibutylamine, and water.
- 12. However, even with Examples 1 and 2, the '826 patent does not teach a method for the preparation of a silicon-containing polysulfide-type polymer by simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond; (B) a polysulfide polymer with

H&H # 071051.00008 Serial No. 10/533,169 at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur, as claimed for the invention. In fact, combining the components of Examples 1 and 2 of the '826 patent would result in a chemical reaction fundamentally different than the reactions utilized in the method of the subject invention.

- 13. Because the chemical reaction utilized in the invention is fundamentally different than the reactions of the '826 patent, the end product formed from each respective reaction is different as well. Thus, the end product formed from the method claimed as the invention is both unexpected and unpredictable in view of the teachings of the '826 patent.
- 14. In particular, in Example 2 of the '826 patent, a blocked polysulfide polymer is reacted with paraformaldehyde in the presence of triethylamine to form a blocked polysulfide polymer. The polysulfide polymer is then reacted with elemental sulfur and catalyzed by n-dibutylamine and water. If Example 2 were to be combined with Example 1, as suggested by the Examiner, the vinyl triethoxy silane and the vinyl triethoxy silane polysulfide of Example 1 would react with the water from Example 2 according to the following reaction mechanisms:
- (I) $-Si(OR)_3 + nH_2O \rightarrow -Si(OH)_n(OR)_{3-n} + nROH$
- (II) 2-Si(OH)_n(OR)_{3-n} \rightarrow -Si(OH)_{n-1}(OR)_{3-n}-O-Si(OH)_{n-1}(OR)_{3-n} + H₂O.

In each of the reaction mechanisms (I) and (II) set forth immediately above, the dash (-) in front of the silicon atom illustrates that the silicon atom may be bonded to a carbon atom of a vinyl group, as in the instance in which it is the vinyl triethoxy silane which is reacting with

water, or the silicon atom may be bonded to an atom in a polysulfide polymer, as in the instance in which it is the vinyl triethoxy silane polysulfide which is reacting with water. As known throughout the art, reaction mechanism (I) is commonly referred to as a hydrolysis reaction, and reaction mechanism (II) is commonly referred to as a condensation reaction. Notably, water is not included in the chemical reaction of the method in the invention. As such, there is neither a water-initiated hydrolysis reaction, as represented by reaction mechanism (I) above, nor a condensation reaction of hydrolysates formed from a water-initiated hydrolysis reaction, as represented by reaction mechanism (II) above, in the invention.

- 15. In addition, after closely analyzing the '826 patent, I can find nothing in the '826 patent that teaches the advantages of utilizing an organic base or ammonia in combination with elemental sulfur during the reaction between an a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond and a polysulfide polymer with at least two mercapto groups in one molecule.
- 16. In particular, Example I of the '826 patent reacts a polysulfide polymer having two mercapto groups per molecule and vinyl triethoxy silane. This reaction is catalyzed by 2,2'-azobisisobutyronitrile, which is known in the art as a radical initiator, and is neither an organic base nor ammonia. In addition, the reaction requires a temperature of up to 110 °C. Therefore, after closely analyzing Example 1 of the '826 patent, I can find nothing which

teaches utilizing an organic base or ammonia or elemental sulfur to catalyze a reaction between a polysulfide polymer having two mercapto groups per molecule and vinyl triethoxy silane, let alone the utilization of an organic base or ammonia and elemental sulfur to catalyze such a reaction, as used in the invention.

- 17. Further, I recognize that Examples 3 and 4 of the '826 patent combine the products formed in Examples 1 and 2. However, in Examples 3 and 4 of the '826 patent, the reactants utilized in Examples 1 and 2 are no longer present due to each respective reaction of Examples 1 and 2. Thus, in Examples 3 and 4 of the '826 patent, there is neither an organic base or ammonia or elemental sulfur individually, let alone a combination of an organic base or ammonia and elemental sulfur, as used in the invention.
- 18. After closely analyzing the '331 patent, I can find nothing in the '331 patent that teaches a method for the preparation of a silicon-containing polysulfide-type polymer by simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond; (B) a polysulfide polymer with at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur, as claimed for the invention.
- 19. Further, after closely analyzing the '131 patent, I can find nothing in the '131 patent, even when analyzed in view of the '331 patent, that teaches a method for the preparation of a silicon-containing polysulfide-type polymer by simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with

H&H # 071051.00008 Serial No. 10/533,169 an aliphatic unsaturated bond; (B) a polysulfide polymer with at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur, as claimed for the invention.

- 20. I recognize that the '331 patent discloses a moisture curable sealant composition which comprises the reaction product of a polysulfide and a silane, where a catalyst, such as pyridine or sulfur, may be utilized. However, upon closely analyzing the teachings of the '331 patent, I would have no reason whatsoever to utilize both an organic base or ammonia and sulfur, nor would I expect the unique advantages associated with utilizing both an organic base or ammonia and sulfur in combination, as in this invention.
- 21. Similarly, I recognize that Example 9 of the '131 patent discloses a process of forming a silyl containing polysulfide polymer by mixing a polysulfide with sulfur and methylvinyldimethoxysilane in a nitrogen atmosphere. However, upon closely analyzing the teachings of the '131 patent, I would have no reason whatsoever to utilize the organic base taught by the '331 patent in Example 9 of the '131 patent such that both an organic base and sulfur are utilized, nor would I expect the advantages of utilizing both an organic base and sulfur in combination, as claimed for the invention.
- 22. The advantages of utilizing the organic base or ammonia in combination with sulfur to catalyze the reaction between a polysulfide polymer having two mercapto groups per molecule and vinyl triethoxy silane are exemplified in Examples 1-3 when compared with Comparative Examples 1 and 2 of the subject application.

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23. In particular, Comparative Examples 1 and 2 each correspond with the teachings of the '331 patent. For example, the '331 patent teaches that pyridine or sulfur may be used as a catalyst. In Comparative Example 1, a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond and a polysulfide polymer with at least two mercapto groups in one molecule were mixed with an organic base. Notably, the reaction between the silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond and the polysulfide polymer with at least two mercapto groups in one molecule did not occur. Similarly, in Comparative Example 2, the silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond and the polysulfide polymer with at least two mercapto groups in one molecule were mixed with elemental sulfur. Once again, the reaction between the silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond and the polysulfide polymer with at least two mercapto groups in one molecule did not occur. Thus, Comparative Example 1 corresponds to the teaching of the '331 patent relating to pyridine, as pyridine is an organic base, and Comparative Example 2 corresponds to the teaching of the '331 patent relating to sulfur. Notably, the organic base and the sulfur were undesirable when used individually, as illustrated in Comparative Examples 1 and 2, and there is no reason to expect the excellent properties obtained by utilizing both an organic base and sulfur based on the teachings of the '331 patent.

24. In addition, Comparative Example 2 corresponds to the teachings of Example 9 of the '131 patent. In particular, as set forth above, Example 9 of the '131 patent discloses a process of forming a silyl containing polysulfide polymer by mixing a polysulfide with sulfur and methylvinyldimethoxysilane in a nitrogen atmosphere. Similarly, in Comparative Example 2, vinyltrimethoxylsilane, diethoxymethane disulfide polymer having molecular terminals capped with mercapto groups, and sulfur are reacted. Thus, three similar components are mixed in Example 9 of the '131 patent and in Comparative Example 2 of the subject application. Notably, in the absence of the organic base, the end product was undesirable for the purposes of this invention, and there is no reason to expect the excellent properties obtained by utilizing both an organic base and sulfur based on the teachings of the '131 patent. Further, there is no reason whatsoever to combine the '331 patent with the '131 patent such that both an organic base and sulfur are utilized, nor is there any reason whatsoever to expect the advantages of utilizing both an organic base and sulfur.

25. Therefore, I believe that after closely analyzing the '826, '331 and '131 patents, one of skill in the art, such as myself, would have no reason whatsoever to react a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond, a polysulfide polymer with at least two mercapto groups in one molecule, and an organic base or ammonia in the presence of sulfur, as claimed as the invention.

Conclusion

26. As a result of my review of the '826 patent and also as a result of my understanding from a perspective of a person skilled in the art, the invention as claimed at the time of filing the patent application presented, and still presents, significant differences from the '826, '331 and '131 patents, as well as from a combination of the '331 and '131 patents. Specifically, none of these references individually, nor combinations of these references, teach a method of preparing a silicon-containing polysulfide-type polymer by simultaneously mixing (A) a silicon-containing compound having a silicon atom-bonded monovalent organic group with an aliphatic unsaturated bond; (B) a polysulfide polymer with at least two mercapto groups in one molecule; and (C) an organic base or ammonia in the presence of (D) sulfur, as in the invention. Further, the advantages of utilizing an organic base or ammonia in combination with sulfur are unexpected based upon the teachings of the '826, '331 and '131 patents, even when these patents are analyzed in combination.

27. In addition, the '826, '331 and '131 patents only broadly disclose each of components (A)-(D). However, in view of the teachings of the '826 patent, I would have no reason whatsoever to pick particular reactants disclosed in the Examples of the '826 patent, while excluding several other reactants also disclosed and essential to the reactions of these Examples to arrive at this invention. In particular, I would have no reason whatsoever to select vinyl triethoxy silane and a polysulfide polymer from Example 1 of the '826 patent, while excluding 2,2'-azobisisobutyronitrile. Further, I would have no reason whatsoever to select triethylamine and elemental sulfur from Example 2 of the '826 patent, while excluding

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water and the step of reacting the polysulfide polymer with paraformaldehyde, and mixing the

triethylamine and elemental sulfur with the vinyl triethoxy silane and the polysulfide polymer

of Example 1 to form a silicon-containing polysulfide-type polymer. Further, in view of the

teachings of the '331 patent, either individually or in combination with the teachings of the

'131 patent, I would have no reason to utilize an organic base or ammonia in combination

with sulfur, nor would I have any reason to expect the advantages of utilizing an organic base

or ammonia in combination with sulfur, as illustrated in Comparative Examples 1 and 2 of

this application.

28. I hereby declare that all statements made herein of my own knowledge are true

and that all statements made on information are believed to be true, and further that these

statements were made with the knowledge that willful and false statements and the like are

punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United

States Code, and that such willful false statements may jeopardize the validity of the

application or patent issued thereon.

Respectfully submitted,

otollwar

Dated

Makoto Iwai

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7, July, 2009

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